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Synthesis and Property Evaluation of Y-Ba-Cu-O Superconductors

by

A.S. Rao
O.P. Arora

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CONTENTS

	Page
ABSTRACT	1
ADMINISTRATIVE INFORMATION	1
INTRODUCTION	1
EXPERIMENTAL PROCEDURE	2
RESULTS	6
DISCUSSION	17
CONCLUSION	18
REFERENCES	21

FIGURES

	Page
1. The flow diagram of the processing of yttrium, barium and copper oxide powder by solid state reaction, chemical coprecipitation and copper oxide coating methods.	3
2. Schematic diagram of the powder synthesis unit	4
3. Scanning electron micrograph of the sintered $\text{YBa}_2\text{Cu}_3\text{O}_{6+x}$ sample morphology. Powder preparation (A) solid state reaction, (B,C) chemical coprecipitation and (D) copper oxide coating with oxalate crystallites of yttrium and barium method.	9
4. Orthorhombic crystal phase versus the superconducting transition temperature (T_c) for samples that were calcined and annealed ($\bigcirc \bullet \square \Delta \nabla$) once and ($\bullet \bullet \blacksquare \blacktriangle \blacktriangledown$) two times. Powder preparation ($\square \square$) solid state reaction, ($\Delta \blacktriangle \nabla \blacktriangledown$) chemical coprecipitation as ($\Delta \blacktriangle$) oxalate and ($\nabla \blacktriangledown$) carbonate of Y-Ba-Cu, and ($\bigcirc \bullet \bullet \bullet$) copper oxide coating as ($\bigcirc \bullet$) oxalate and ($\bullet \bullet$) carbonate of Y-Ba, method.	11



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5. Electrical resistivity of sintered $\text{YBa}_2\text{Cu}_3\text{O}_{6+x}$ samples produced by solid state reaction method. Samples were calcined at 940°C and were annealed at 550°C (○) once and (●) two times. 12
6. Electrical resistivity of sintered $\text{YBa}_2\text{Cu}_3\text{O}_{6+x}$ samples produced by chemical coprecipitation method as oxalates. Samples were calcined at 920°C and were annealed at 550°C (□) once and (■) two times. 13
7. Electrical resistivity of sintered $\text{YBa}_2\text{Cu}_3\text{O}_{6+x}$ samples produced by chemical coprecipitation method as carbonates. Samples were calcined at 920°C and were annealed at 550°C (△) once and (▲) two times. 14
8. Electrical resistivity of sintered $\text{YBa}_2\text{Cu}_3\text{O}_{6+x}$ samples produced by copper oxide coating method with Y and Ba oxalates. Samples were calcined at 920°C and were annealed at 550°C (○) once and (●) two times. 15

TABLES

	Page
1. Yttrium barium and copper oxide precursor powder characteristics.	7
2. Yttrium, barium and copper oxide powder characteristics. Powder calcination and annealing temperatures were 920 and 550°C respectively. Solid state reaction temperature and recalcination temperature was 940°C	7
3. Normal state electrical resistivity measured at 100 K of Y-Ba-Cu-O superconductor processed by solid state reaction, chemical coprecipitation and copper oxide coating methods.	16

ABSTRACT

Superconducting $\text{YBa}_2\text{Cu}_3\text{O}_{6+x}$ powders were synthesized for obtaining fine particles of uniform size using solid state chemical reaction, coprecipitation and copper oxide coating with yttrium and barium oxalate or carbonate methods. The powders obtained from all three different routes showed superconductivity; however the T_c values for the as synthesized powders via. the solid state and copper oxide coating methods were ~ 90 K and the oxalate and carbonate derived powders was ~ 80 and 60 K respectively. Recalcination and annealing the samples in oxygen had very little effect on the T_c value, but the sample resistance above T_c appears to decrease slightly.

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INTRODUCTION

The recent surge of interest in understanding the basic physics and chemistry of high temperature ceramic superconductors, after the report of Bednorz and Muller [1] followed by other researchers [2-7], has opened a new era for the development of a different kind of technology. Although in the open literature there have been many claims for the processing of these materials into different shapes,

many engineering properties of these materials have yet to be determined. As a part of an effort to process these ceramic superconductors into long extruded wires using the superplastic deformation, this investigation was undertaken. In this paper, we present the results of the synthesis of fine superconducting $\text{YBa}_2\text{Cu}_3\text{O}_{6+x}$ powder obtained by chemical precipitation, and the results obtained on the powder characteristics and the properties of the sintered materials as a function of process methodology.

EXPERIMENTAL PROCEDURE

Yttrium, barium and copper oxide powders were prepared by solid state chemical reaction, chemical coprecipitation and copper oxide coated powder methods. The flow diagram of the processing is shown in Figure 1. In the solid state method the oxides of yttrium and copper and the carbonate of barium were mixed in predetermined quantities. The mixture was later ball milled in a minimum amount of distilled water. The mixture was dried and was calcined in air. In the coprecipitation method, nitrates of yttrium, barium and copper were dissolved in distilled water. The three solutions were mixed and was stirred for two hours.

Unlike the conventional coprecipitation method, a slightly different precipitation procedure was adopted in our process; viz. using a special spray atomization unit that was designed and built exclusively for the production of fine and narrow size droplets, the solution was sprayed into a pool of either oxalate or carbonate solution that was continuously stirred. A schematic diagram of the spray atomization unit is shown in Figure 2. The spray atomization unit consists of a pressure vessel fitted with a special nozzle at

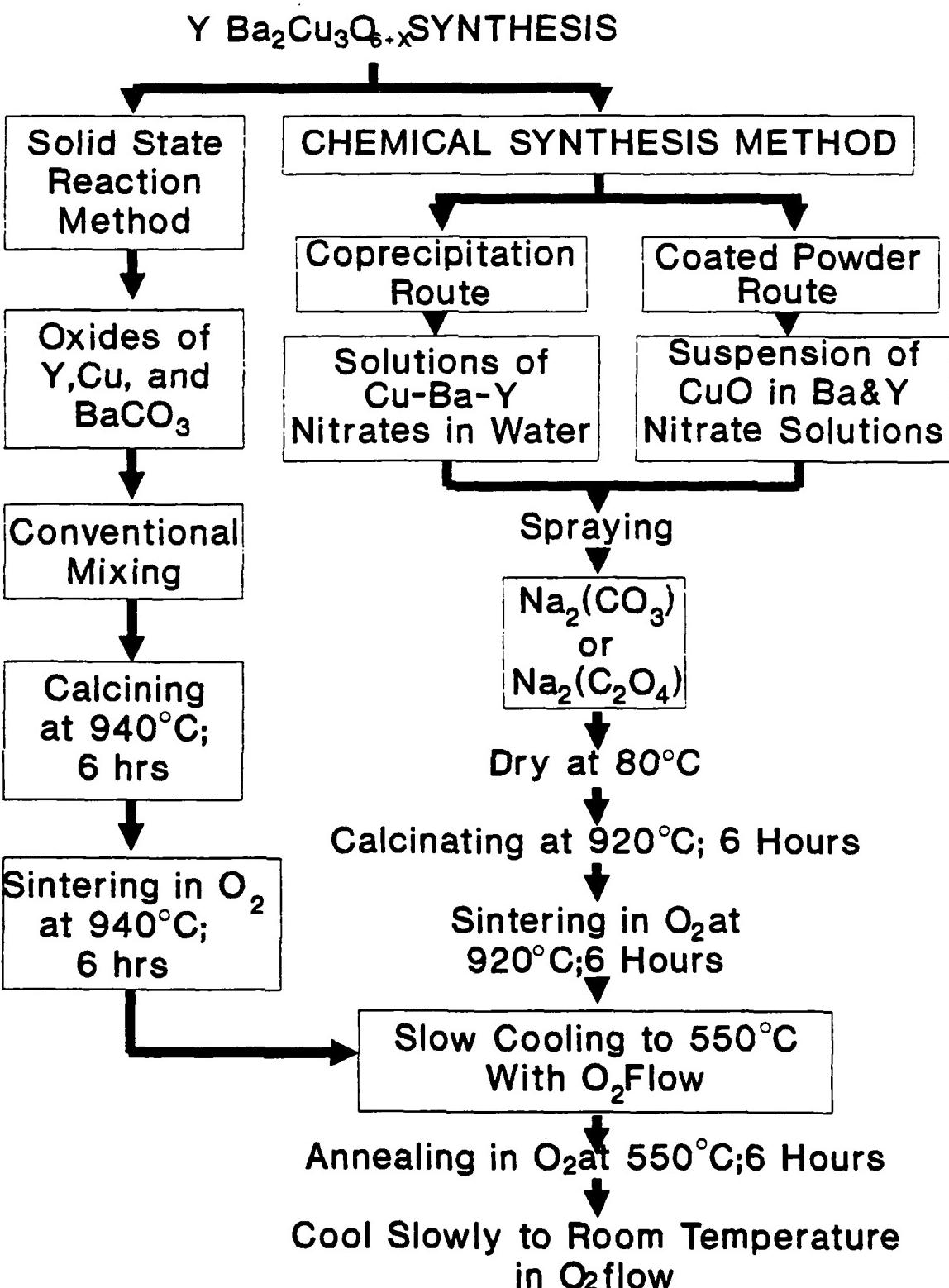


Figure 1. The flow diagram of the processing of yttrium, barium and copper oxide powder by solid state reaction, chemical coprecipitation and copper oxide coating methods.

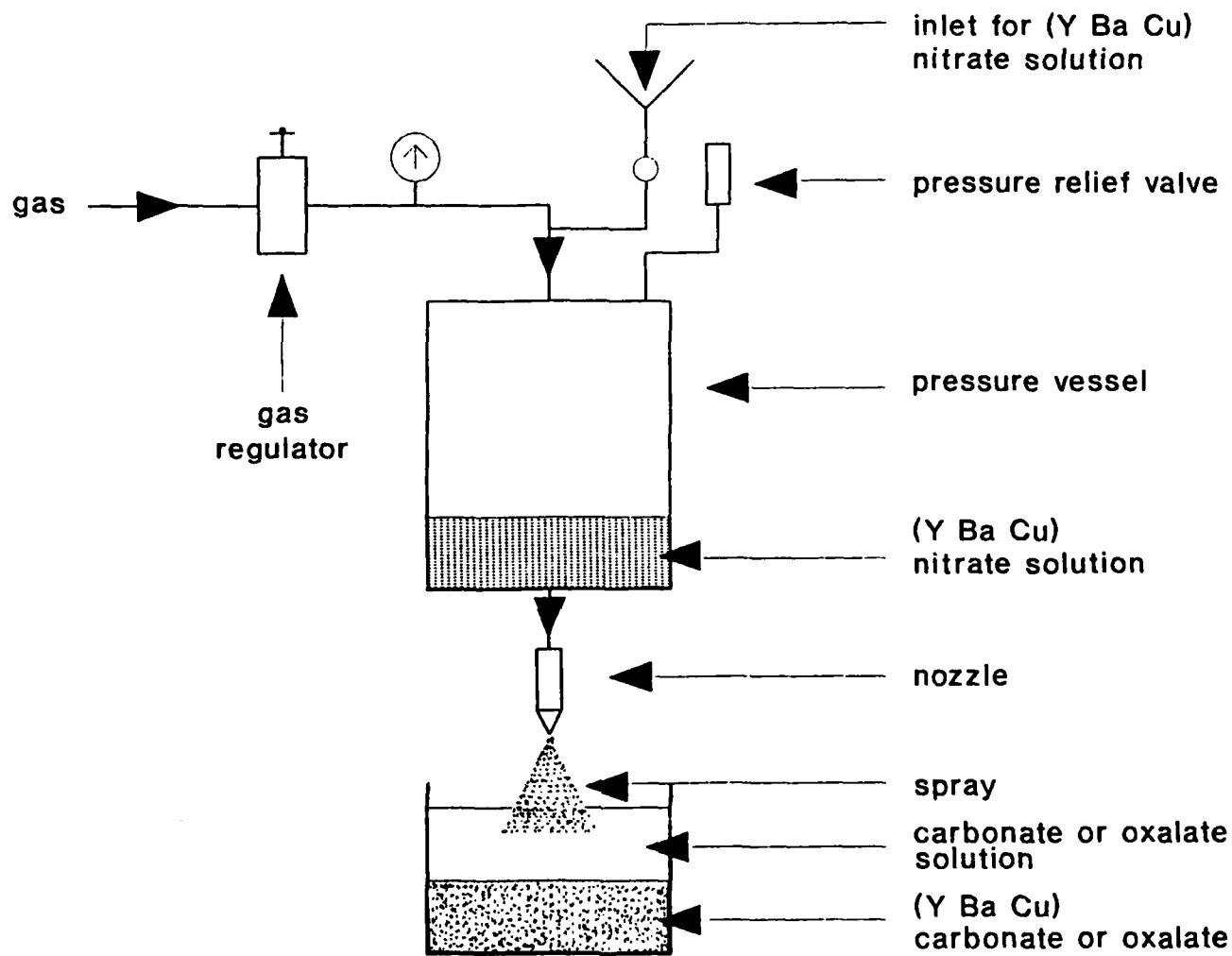


Figure 2. Schematic diagram of the powder synthesis unit

the bottom and an air inlet on the top. The unit was designed so that an application of gas pressure of 100 psi on the top, produces an uniform pressure of ~ 10,000 psi at the exit. It was found from oxide and mixed oxide synthesis that the droplets leaving the spray unit forms solid particles instantaneously upon impact with the surface of the pool of the coprecipitation medium. It was also noticed that, since the nucleated solid particles still maintain nearly the same droplet velocity (after impact), the particles tend to dive deep into the precipitation medium and settle as individual particles but not as agglomerated clumps. The powder in suspension was centrifuged at 4000 rpm for one hour. The clear filtrate was discarded. The solids were redispersed in acidified water (pH ~ 4) and the contents were centrifuged. The dispersion and centrifugation was repeated a few times in order to ensure the removal of excess carbonate or oxalate solution. Later the wet solid was dried in an oven overnight at 80°C and was calcined at 920°C in air.

During processing, particularly the coprecipitation of yttrium, barium and copper ions as carbonates, it was found that the carbonate of copper tends to redissolve in the dispersion medium (distilled water) if the starting solution is too dilute. However, if the nitrate and / or the suspension concentration is too high, the nucleated small crystallites tend to flocculate and settle as large agglomerated clumps. In order to overcome the problem of copper dissolution, a new approach was adopted, similar to that which we used on alumina / zirconia system [8]. In this process, a copper oxide and yttrium and barium nitrate suspension was prepared.

The suspension was sprayed into a pool of oxalate or carbonate solution. The consolidation and calcination procedures for this processin, were similar to that described above. Since the copper oxide particle surface was coated with yttrium and barium nitrate solution, upon precipitation each copper oxide particle surface was found to be saturated with fine crystallites of yttrium and barium oxalate or carbonate.

All calcined $\text{YBa}_2\text{Cu}_3\text{O}_{6+x}$ samples except the samples produced from the solid state chemical reaction process, crumbled into a fine powder upon gentle grinding. The samples were divided into two batches. The first batch of these powders was used for the determination of powder characteristics, such as the density, chemical composition, particle size and surface area. A few 1 cm discs were dry pressed from the second batch of powder samples. These discs were sintered at 920°C in oxygen and were annealed at 550°C in air. The samples containing $\text{YBa}_2\text{Cu}_3\text{O}_{6+x}$ powder produced from solid state chemical reaction were sintered at 940°C and were annealed at 550°C in oxygen. The sintered discs were used for the resistivity measurements, X - ray diffraction and SEM analysis.

RESULTS

The $\text{YBa}_2\text{Cu}_3\text{O}_{6+x}$ powder characteristics and the properties of the as - synthesized oxalate and / or carbonate derived powders are shown in Tables 1 and 2 respectively. These results suggest that the grain size of the yttrium, barium and copper oxide particles obtained from solid state reaction method is larger than that of the powders synthesized from other processing methods. The results also

Table 1. Yttrium barium and copper oxide precursor powder characteristics.

Precursor Powder Characteristics	Coprecipitation Method		Copper Oxide Coating Method	
	Oxalate	Carbonate	Oxalate	Carbonate
Powder Density (gm/cc)	4.9	4.6	4.6	4.6
Surface Area (m^2/gm)	85	80	26	22
Mean Crystallite Size estimated from Surface Area of the Powder (nm)	10	12	60	66

Table 2. Yttrium, barium and copper oxide powder characteristics. Powder calcination and annealing temperatures were 920 and 550°C respectively. Solid state reaction temperature and recalcination temperature was 940°C.

Powder Characteristics	solid state reaction Method	Coprecipitation Method		Copper-Oxide Coating Method	
		Oxalate	Carbonate	Oxalate	Carbonate
Powder density (gm/cc)	6.2	6.0	5.6	5.8	5.7
Surface Area (m^2/gm)	0.2	1.6	1.8	1.2	1.3
Particle Mean diameter (microns)	~10	~1.0	~1.6	~2.0	~2.6
Bulk density of the sintered material (% theoretical)	92	90	83	92	88

suggest that the powder density and the sintered material bulk density of the solid state reaction method is higher than the other materials. It has to be emphasized that although the sintered sample obtained from solid state reaction has a higher density, the differences many not represent the true nature of the process methodology. It is because while the sintering temperature for solid state reaction powders was 940°C the other powders were sintered at 920°C. We avoided a higher sintering temperature of 940°C for powders derived from the chemical precipitation methods, because the fine oxalate or carbonate or copper oxide coated precursors showed a considerable melting when heated above 930°C. For solid state reaction, we noticed that the chemical reaction is incomplete for calcination below 930°C.

The morphology of the sintered $\text{YBa}_2\text{Cu}_3\text{O}_{6+x}$ ceramic obtained from all four processing methods is shown in Figure 3. The results suggest that while the sintering process produced long elongated platelet like grains for solid state reaction, copper oxide coated powder and coprecipitation via. oxalate precursor methods, the carbonate derived powder tends to sinter into nearly square shaped discs and large porosity. The structure of all $\text{YBa}_2\text{Cu}_3\text{O}_{6+x}$ ceramic samples was determined using wide angle X-ray diffraction method. Some of our samples were analyzed using a Guinier focusing camera. We found that it was very useful to obtain Guinier X-ray plots on all our $\text{YBa}_2\text{Cu}_3\text{O}_{6+x}$ samples because these plots are very sensitive in detecting even small concentration of tetragonal crystal structures in a predominantly orthorhombic structure matrix. From both the Guinier X - ray plots and X- ray diffraction plots, the

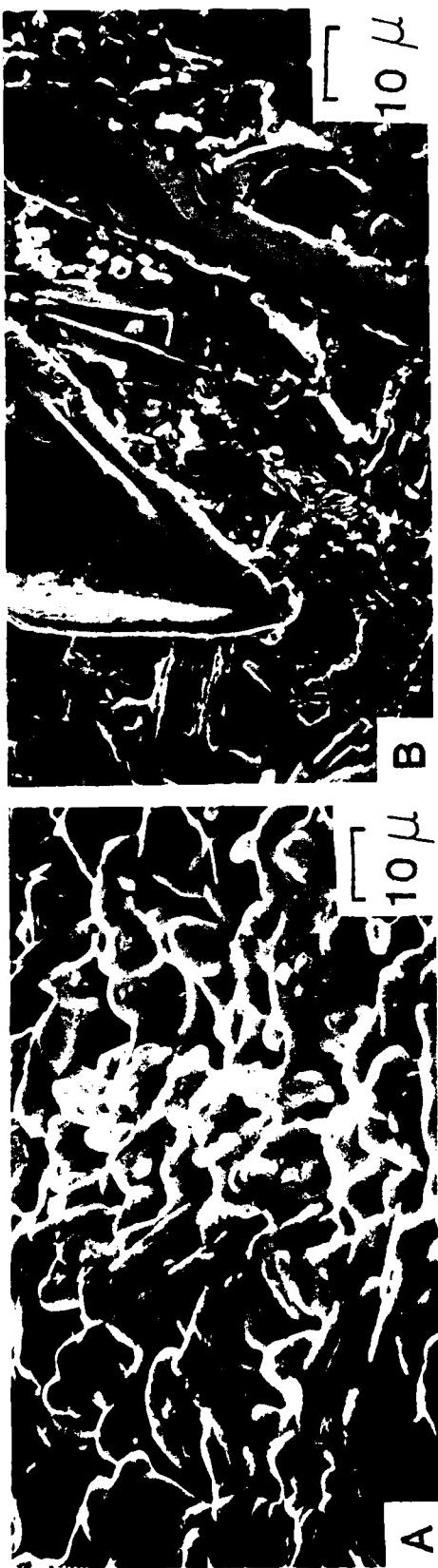


Figure 3. Scanning electron micrograph of the sintered $\text{YBa}_2\text{Cu}_3\text{O}_{6+x}$ sample morphology. Powder preparation (A) solid state reaction, (B,C) chemical coprecipitation and (D) copper oxide coating with oxalate crystallites of yttrium and barium method.

percentage of the orthorhombic phase of all samples was determined (Figure 4). These results suggest that the yttrium, barium and copper oxide powders produced from the solid state reaction and the copper oxide coating process retained the maximum orthorhombic crystal structure. The carbonate precursor derived powder, on the other hand, retained the minimum orthorhombic crystal phase. From Figure 4, it can also be noted that recalcining and annealing the samples in oxygen, except for the carbonate derived material, did not improve the % orthorhombic phase of the original sample.

Electrical resistivity of samples obtained from solid state reaction, coprecipitation and copper oxide coating methods is shown in Figures 5 - 8 respectively. The normal state resistivity of these samples measured at 100 K is shown in Table 3. The yttrium, barium and copper oxide produced from solid state reaction and copper oxide coating methods show nearly the same superconducting transition temperature (T_c) at ~ 90K, but their normal state resistivity varied considerably. The samples obtained via coprecipitation as oxalate or carbonate showed superconducting behavior (T_c) below ~80 and 60 K respectively. However, the T_c of carbonate derived powder has increased to ~ 70 K upon recalcination and annealing in oxygen.

In order to derive some semi- quantitative correlation between the superconducting transition temperature (T_c) and the superconducting orthorhombic crystal structure, the % orthorhombic phase (obtained from x-ray analysis) for all $\text{YBa}_2\text{Cu}_3\text{O}_{6+x}$ samples was plotted against the T_c obtained from electrical resistivity measurements. The results (Figure 4) suggest that for the

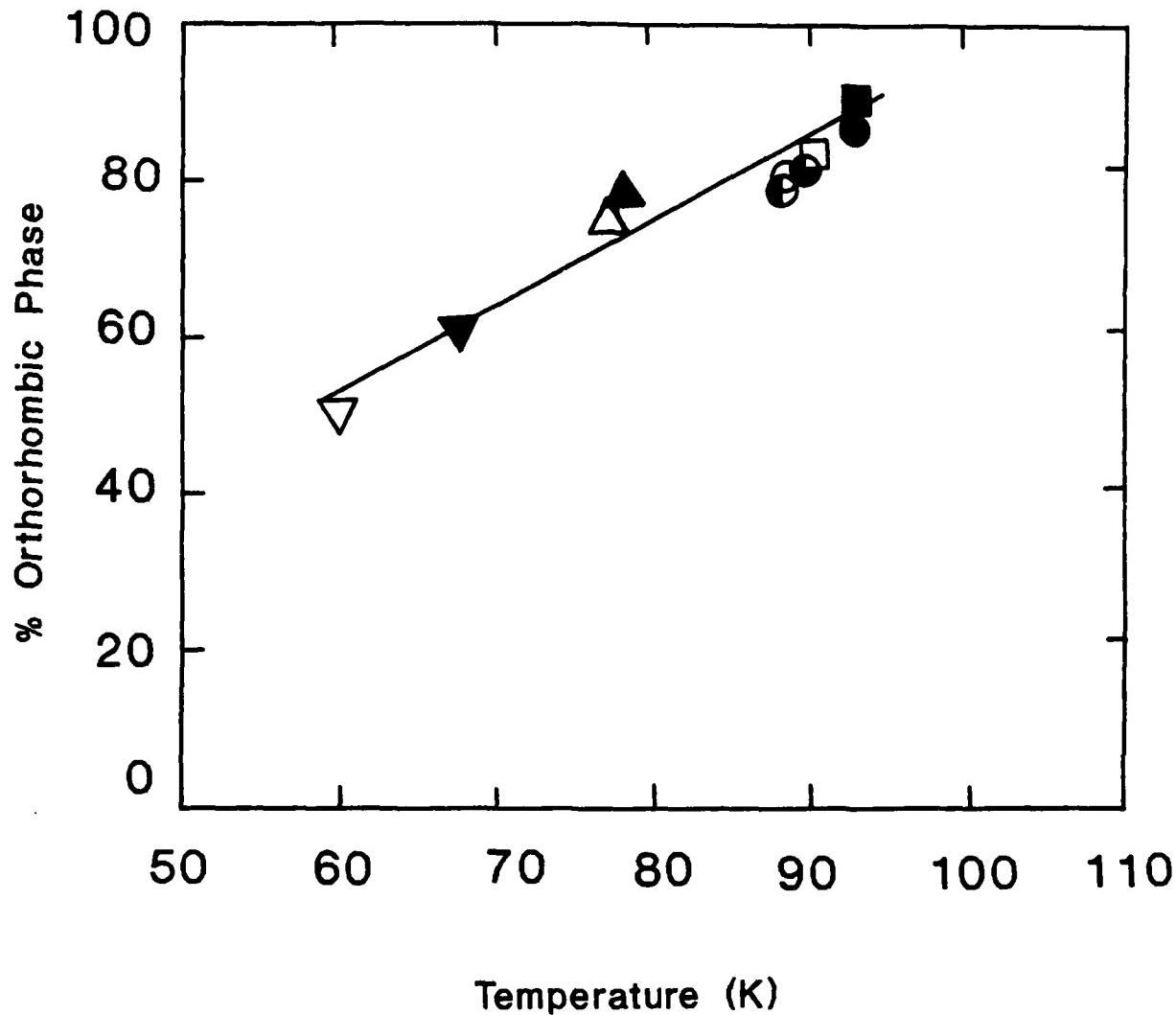


Figure 4. % Orthorhombic crystal phase versus the superconducting transition temperature (T_c) for samples that were calcined and annealed (○□△▽) once and (●○■▲▼) two times. Powder - preparation (□■) solid state reaction, (△▲▽▼) chemical coprecipitation as (△▲) oxalate and (▽▼) carbonate of Y-Ba-Cu, and (○●○●) copper oxide coating as (○●) oxalate and (●●) carbonate of Y - Ba, method.

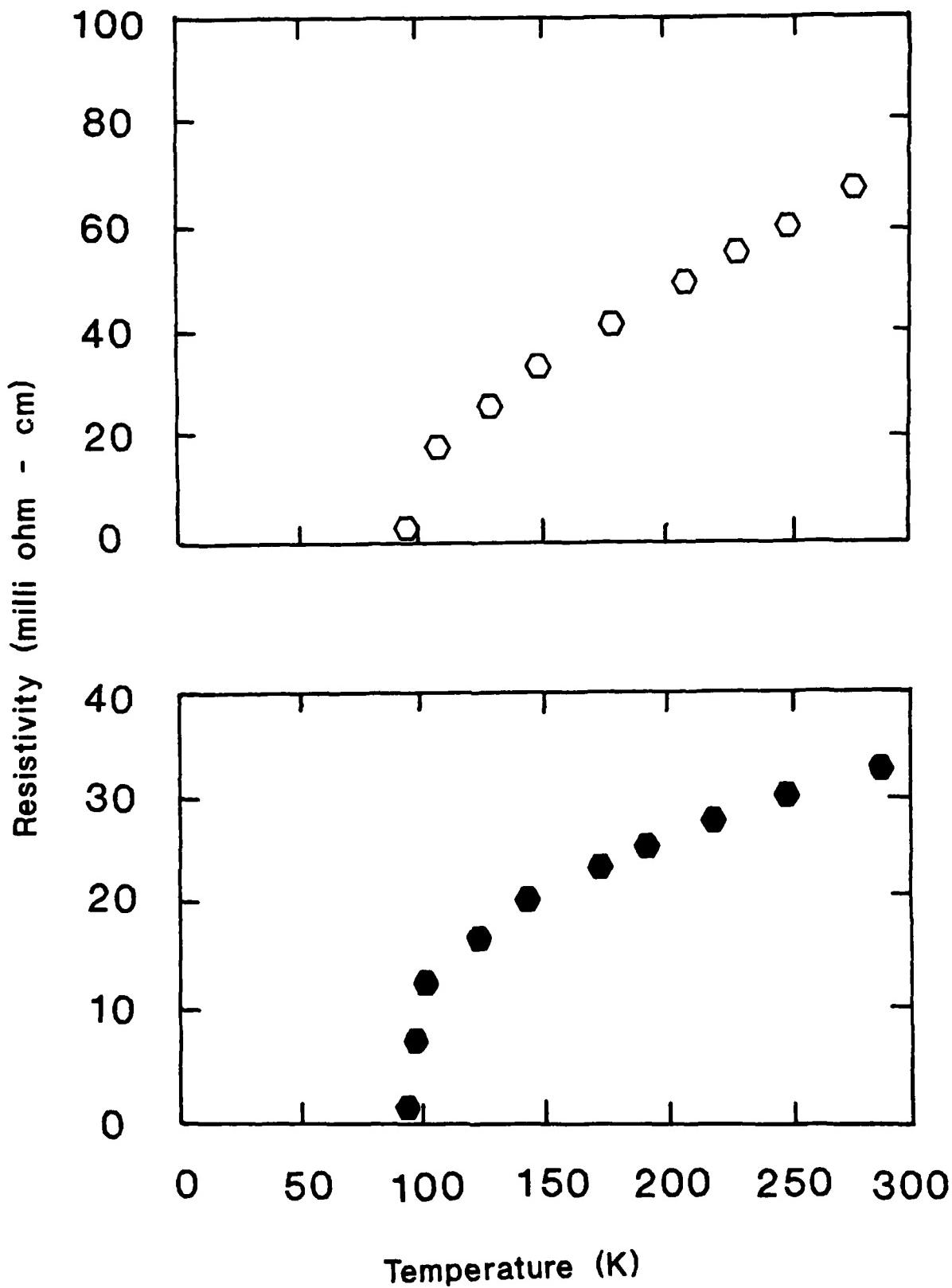


Figure 5. Electrical resistivity of sintered $\text{YBa}_2\text{Cu}_3\text{O}_{6+x}$ samples produced by solid state reaction method. Samples were calcined at 940°C and were annealed at 550°C (○) once and (●) two times.

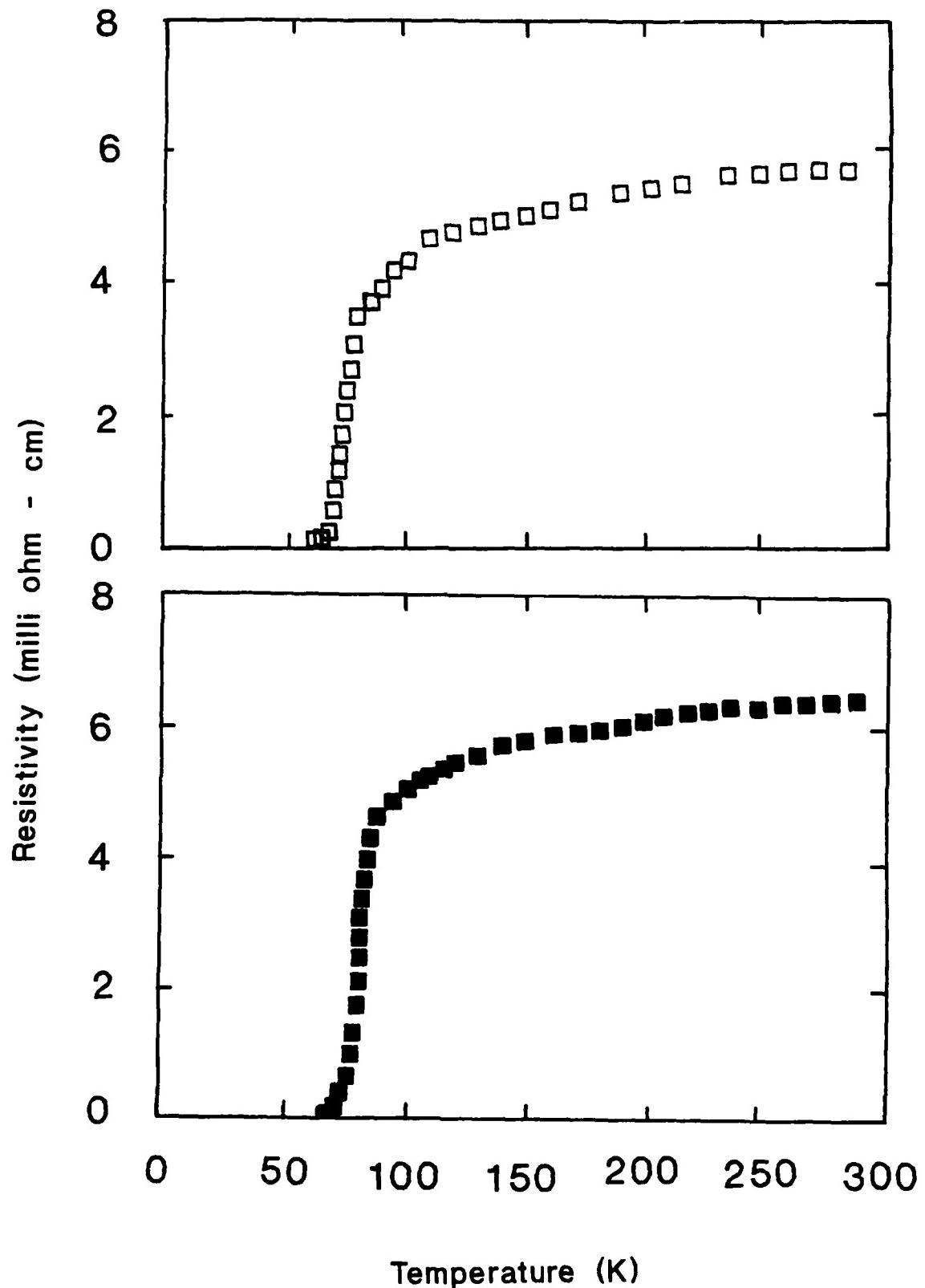


Figure 6. Electrical resistivity of sintered $\text{YBa}_2\text{Cu}_3\text{O}_{6+x}$ samples produced by chemical coprecipitation method as oxalates. Samples were calcined at 920°C and were annealed at 550°C (□) once and (■) two times.

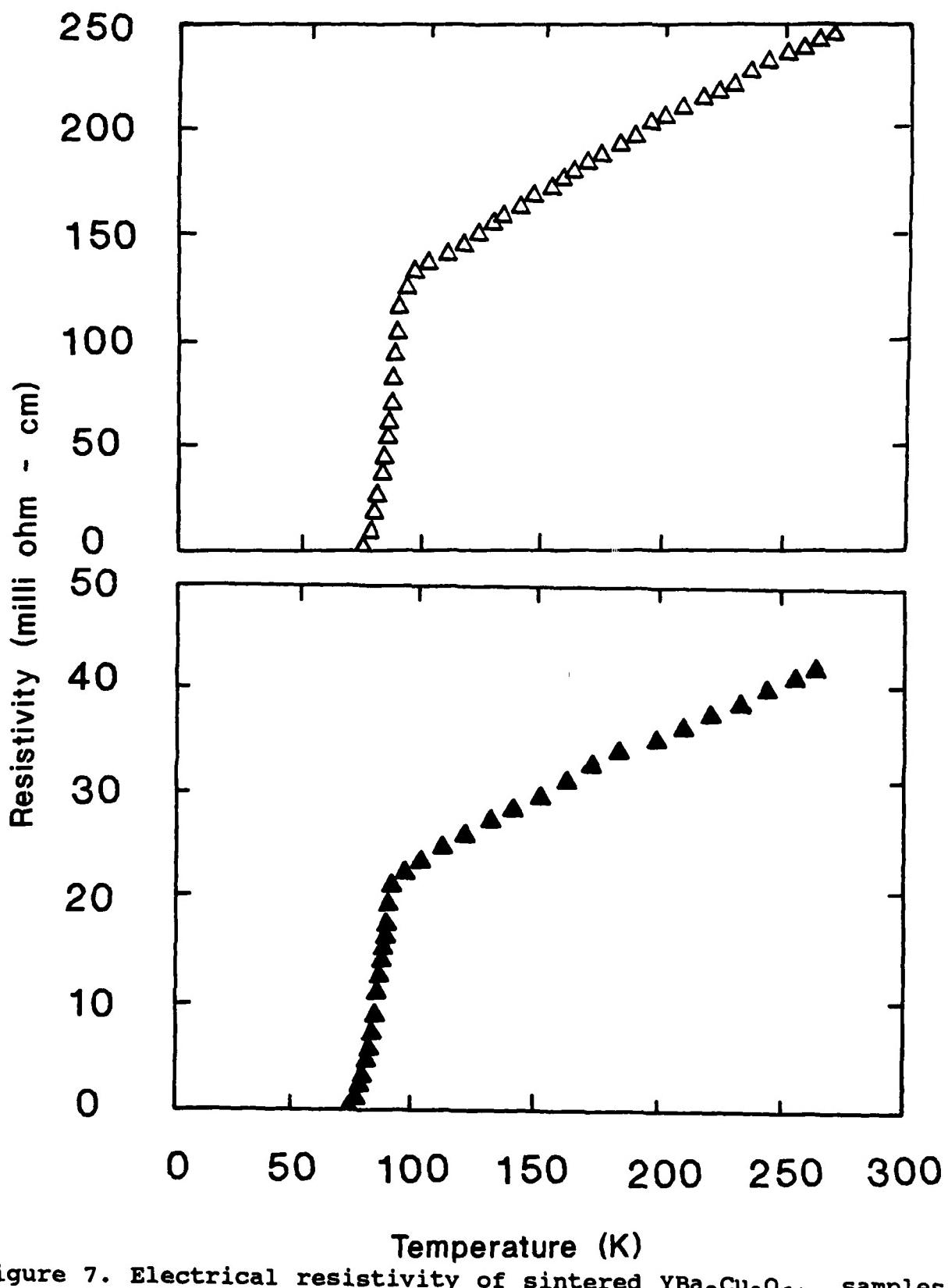


Figure 7. Electrical resistivity of sintered $\text{YBa}_2\text{Cu}_3\text{O}_{6+x}$ samples produced by chemical coprecipitation method as carbonates. Samples were calcined at 920°C and were annealed at 550°C (Δ) once and (\blacktriangle) two times.

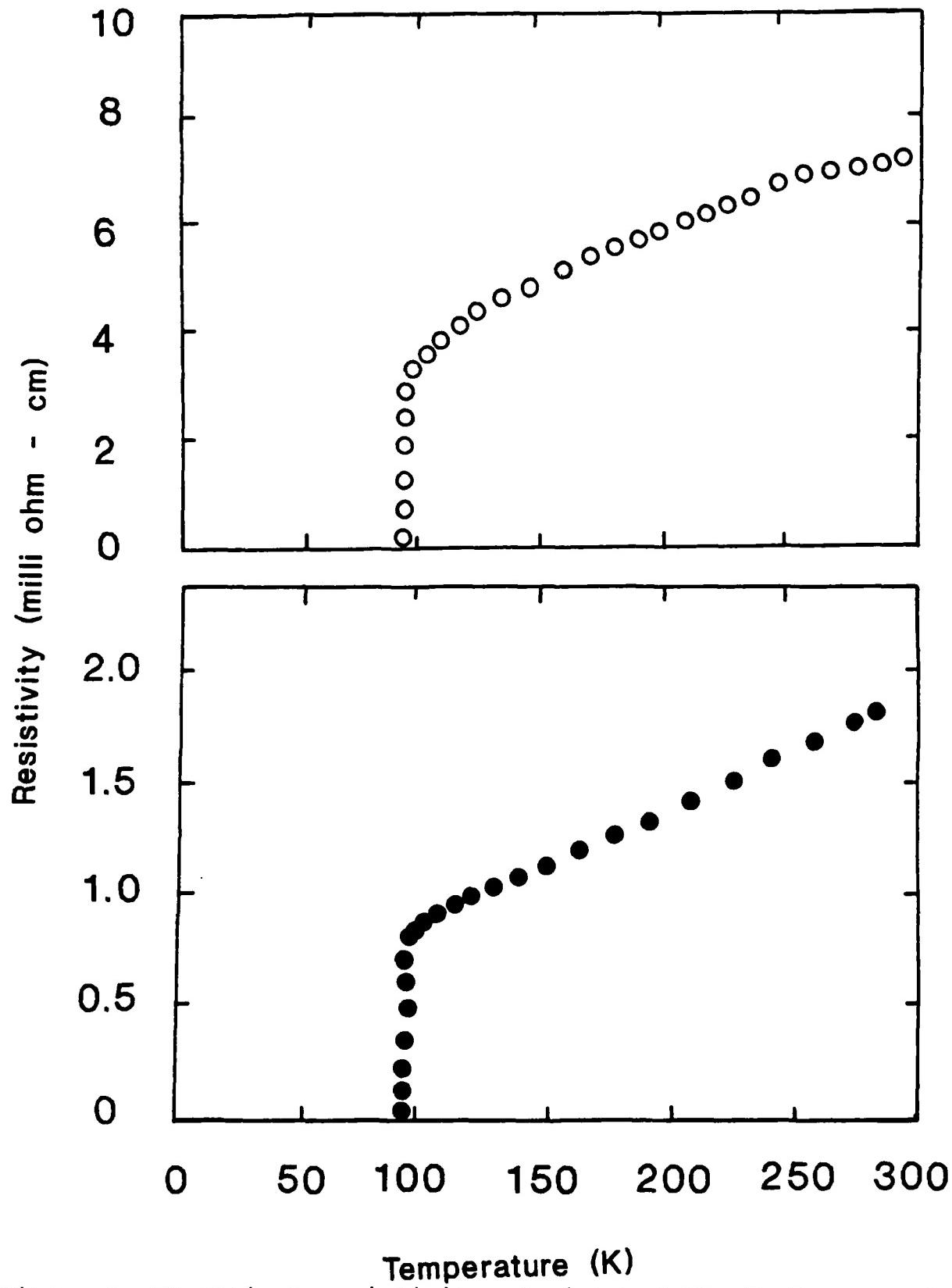


Figure 8. Electrical resistivity of sintered $\text{YBa}_2\text{Cu}_3\text{O}_{6+x}$ samples produced by copper oxide coating method with $\text{Y} - \text{Ba}$ oxalates. Samples were calcined at 920°C and were annealed at 550°C (○) once and (●) two times.

Table 3. Normal state electrical resistivity measured at 100 K of Y-Ba-Cu-O superconductor processed by solid state reaction, chemical coprecipitation and copper oxide coating methods.

Processing Methodology	No. of Calcination and Annealing Cycles	Superconducting Transition Temp. (K)	Normal State Electrical Resistivity (milli ohm-cm)
Solid State	1	91	20
Reaction Method	2	93	12
Coprecipitation Method (Via Oxalate Route)	1	78	5
	2	80	4.5
Coprecipitation Method (Via Carbonate Route)	1	61	125
	2	69	25
Copper Oxide Coating Method (Via Oxalate Route)	1	89	3
	2	93	0.7
Copper Oxide Coating Method (Via Carbonate Route)	1	86	5
	2	90	1.5

superconducting powders investigated here, the T_c increases linearly with an increase in the % orthorhombic crystal phase of the superconductor. The results also indicate that the T_c does not depend upon the pre- processing history of the superconductor (viz. the process methodology or the precursor chemistry etc.). We have also observed a similar trend in $\text{YBa}_2\text{Cu}_3\text{O}_{6+x}$ superconducting ceramic material containing up to 0 - 30 wt.% of either alumina or silver oxide [9].

DISCUSSION

It has been established from a number of superplastic deformation studies of both metallic and ceramic materials, that for maximum deformation it is important that the grain size be as small as possible; typically 1000 - 5000 Å. Although, our present process did not produce such fine sintered particle sizes, it has demonstrated that a careful control of the sintering process of fine crystallites that were made by spray atomization during sintering can produce fine grained material. It is interesting to note from the results how complex the sintering of these materials is. For example, the carbonate precursor powders sinter into nearly square shaped discs while the oxalate or copper oxide coating precursor powder tends to sinter into long elongated platelets. It would also be interesting to determine why the carbonate derived material retains more of a tetragonal phase than the material produced from other precursors.

Except for the solid state chemical reaction method where the as-received yttrium, copper oxide and barium carbonate were

chemically reacted without any particle size control, both coprecipitation process and copper oxide coating method involved, first the production of fine 100 Å crystallites. A simple mechanism for the crystallite transformation during calcination can be suggested as follows : The fine 100 Å crystallites during the calcination process first decompose into their respective oxides, which then react to form mixed yttrium, barium and copper oxide ($\text{YBa}_2\text{Cu}_3\text{O}_{6+x}$). Later the fine oxide particles sinter and form a sintered superconducting bulk ceramic. Similar results have been reported in the literature for sintering of other ceramic materials such as hydrous zirconia crystallites in zirconia alumina composites [7]. The mechanism that was suggested for fine crystallites growth during sintering is as follows.

During the calcination process the hydrous zirconia first decomposes to produce pure zirconia crystallites. These crystallites then migrate into the vicinity of each other and coalesce to form large zirconia grains of ~ 0.5 microns. Since the yttrium, barium and copper carbonate or oxalate calcination process involves a similar decomposition and grain growth process, it is reasonable to expect that the final material microstructure to be nearly the same. Since our results indicate that the microstructures depend upon the precursor material, it is reasonable to suggest that the chemical synthesis of $\text{YBa}_2\text{Cu}_3\text{O}_{6+x}$ and sintering process are interdependent. It therefore requires basic understanding of the fine crystallite surface chemistry as a function of sintering process variables, such the surface forces, energy of attraction and repulsion between fine crystallites. We

intend to address these questions in our on going program and hope to report the results at a later date.

CONCLUSION

From the present investigation the following conclusions can be derived :

1. Fine superconducting yttrium, barium and copper oxide powders can be produced either from chemical coprecipitation or the copper oxide coating method using spray atomization process.
2. While the copper oxide coating process produced the superconducting ceramic with T_c comparable to the powders produced from conventional solid state reaction ($T_c \sim 90$ K), the T_c of the powders produced from coprecipitation process is in the range 60 - 80 K.
3. The normal state resistivity of the powder produced from the copper oxide coating method is the lowest of all the powders investigated.
4. The carbonate precursor derived superconductor not only retained the maximum concentration of tetragonal crystal phase, but also tends to sinter into square shaped discs.
5. The superconducting transition temperature and the normal state resistivity of the sintered $\text{YBa}_2\text{Cu}_3\text{O}_{6+x}$ ceramic does not change significantly due to recalcination and annealing in oxygen.

REFERENCES

1. Bednorz, J. C. and Muller, K. A., "Possible high T_c superconductivity in the barium - lanthanum - copper - oxygen system," *Z. Phys. B. Condensed Matter*, 64, 189 (1986).
2. Wu, M. K., Ashburn, J. R., Tong, C. T., Hor, P. T., Meng, R. L., Cao, L., Huan, Z. J., Wang, Y. Q. and Chu, C. W., "Superconductivity at 93 K in a new mixed phase Y-Ba-Cu-O compound system at ambient pressure," *Phys. Rev. Letters*, 58, 908 (1987).
3. Advanced Ceramic Materials, 2 (1987).
4. Novel Superconductivity, edt. Wolf, S. A. and Kresin, V. Z., Plenum Publ. Co. (1987).
5. High Temperature Superconductivity, edt. Ginzburg, V. L. and Kirzhnits, Plenum Pub. Co., (1987).
6. Chemistry of High Temperature Superconductors, edt. Nelson, D. L., Whittingham, M. S. and George, T. F., Amer. Chem. Soc., (1987).
7. Rao, C. N. R., Ganguly, P., Gopalakrishnan, J. and Sarma, D. D., "Mechanism of high temperature superconductivity in yttrium, barium and copper oxide ($YBa_2Cu_3O_{7-x}$) : Crucial role of oxygen," *Mater. Res. Bull.*, 22, 1159 (1987).
8. Srinivasa Rao, A. and Cannon, W. R., "Alumina / zirconia composites produced from commercial alumina and synthesized hydrous zirconia," *Ceram. International*, 15, 179 (1989).
9. Srinivasa Rao, A., Aprigliano, L. and Arora, O. P., "Effect of additives on the morphology and superconductivity of $YBa_2Cu_3O_{6+x}$ ceramic materials," To be Published in Int. Conf. on Superconductivity, Bangalore, India (1990).

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